

REMARKS

By this amendment, applicants have amended claim 11 to include therein the limitations previously recited in claim 13. Accordingly, claim 13 has been canceled without prejudice or disclaimer.

Claims 1, 6 and 10-14 stand rejected under 35 U.S.C. 103(a) as being unpatentable over Japanese Patent Application Publication Number JP-2001-243951 (Matsumoto et al.) in view of International Application Publication Number WO 03/044881 (Shiozaki et al.). Applicants traverse this rejection and request reconsideration thereof.

The first feature of the present invention is in that “length in which the primary particles are linked on the section of the secondary particle is equivalent to 10 to 70% of the length of the whole periphery on the section of the primary particles.” This feature is based on the fact that the inventors of the present invention had found as described on page 4, lines 1-8 of the specification in English text. That is, the ionic conductivity is deteriorated at low temperature, since the viscosity of electrolytic solution decreases at low temperature. If each secondary particle of positive electrode material is formed with flocculated primary particles and the primary particles are positioned apart from one another so that the spaces among the primary particles are filled with electrolytic solution, a conductive network among the primary particles is locally decreased. Therefore, the resistance increases and the voltage drop of a battery occurs.

It is necessary to maintain the ionic conductivity to prevent the voltage drop of a battery. There are two ways to maintain the ionic conductivity, one being by maintaining the ionic conductivity of lithium in electrolytic solution, the other being by maintaining the diffusion of lithium ions between the primary particles.

The inventors of the present invention have found the fact that at low temperature, the diffusion of lithium ions between the primary particles is more important than the traveling of lithium ions via electrolytic solution, since the ionic conductivity is deteriorated at low temperature. Accordingly, by increasing the contact areas among primary particles, it is possible to maintain the conductive network in the environment of low temperature.

Fig. 2 shows that a secondary particle 2 of positive electrode material is formed with flocculated primary particles 1. For the cross section of each primary particle, the total length of the shared sides with the adjoining primary particles is calculated and compared with the length of the periphery of the primary particle. The desirable average of such contact length compared with the periphery of the section of the primary particle is 10 to 70%.

Generally, volume change in a positive electrode material will occur when lithium ions come into it or go out from it due to charging/discharging. If the volume change in the positive electrode material is relatively large and the contact areas among primary particles are increased, primary particles may be crushed at the contact areas due to the volume change. This causes the life span of the battery to decrease.

The second feature of the present invention is in that the secondary particle is represented as $\text{Li}_a\text{Mn}_x\text{Ni}_y\text{Co}_z\text{O}_2$, and the secondary particle is composed of crystals having layer structure of composite oxide meeting $1 < a \leq 1.2$, $0 \leq x \leq 0.65$, $0.35 \leq y < 0.5$, $0 \leq z \leq 0.65$ and $x + y + z = 1$. In other words, the second feature of the present invention is in that the percentage content of Ni (y) is less than 50%, so that the relative lattice volume change rate decreases (on page 15, lines 23-24).

Fig. 7 shows that if the percentage content of Ni is 50% or less, the change of lattice volume due to charging/discharging decreases. Therefore, it is possible to maintain the high contact areas among the primary particles, and the high capacity maintenance rate.

As described above, there is a trade-off relation between the low temperature performance and the life span of the battery. If the low temperature performance is to be improved, the contact area among the primary particles should be increased to promote the diffusion of lithium ions between primary particles, which causes the life span of battery to decrease. If the life span of battery is to be increased, the contact areas among the primary particles should be decreased to prevent the primary particles from being crushed, but the low temperature performance is deteriorated.

In order to maintain both the low temperature performance and life span of battery, it is necessary to avoid secondary particles that include only such primary particles that the contact areas among the primary particles are small.

The third feature of the present invention is in that the voidage of the secondary particle is 2.5 to 35 %. In order to avoid secondary particles that include only such primary particles that the contact areas among the primary particles are small, the third feature of the invention is necessary. As described in the specification, in English text on page 5, lines 18-24, if the voidage is 2.5% or less, the maintainability of electrolytic solution is deteriorated, the room-temperature characteristic is deteriorated, and, therefore, the voidage for maintaining the rechargeable capacity at room temperature is 2.5% or more. In the meantime, if the voidage is 35% or more, a conductive network is inhibited and the low-temperature characteristic is deteriorated.

By combining the first, second and third features of the present invention, a lithium secondary battery using the positive electrode material in which the discharge rate characteristic and the battery capacity in the environment of the low temperature of -30°C are hardly deteriorated and which are excellent in a cycle characteristic can be achieved.

Japanese Patent Publication 2001-243951 (Matsumoto et al.) discloses a positive electrode active material for a non-aqueous electrolyte secondary battery, which is comprised of secondary particles, each of secondary particle being composed of fine primary particle of lithium cobalt oxide. At least a part of the fine primary particles in a secondary particle are arranged in radiation toward outside from the center of the secondary particles. The secondary particles have many fine gaps among primary particles, in which the electrolyte can infiltrate.

In the case of such a positive electrode active material, the structure at the center of the secondary particle is different from that at the radially outer portion of the secondary particle. At the center of the secondary particle, the gaps among the primary particles are relatively small, and therefore, electron conductivity is maintained. However, at the radially outer portion of the secondary particle, the gaps among the primary particles are relatively large, and therefore, the contact areas among primary particles are small. Therefore, at the radially outer portion of the secondary particle, electron conductivity decreases.

At the radially inner portion of the secondary particle, the contact areas among primary particles are relatively large for the reasons described hereafter.

According to Matsumoto et al., at least a part of primary particles are connected by sintering, in order to increase electron conductivity among primary particles at the radially outer portion of the secondary particle. However, connection of primary particles by sintering proceeds considerably at the center area of the

secondary particle, as compared with that at the radially outer portion of the secondary particle, since gaps among the primary particles are relatively small at the center area of the secondary particle. If the sintering condition is set to be suitable for the connection at the radially outer portion of the secondary particle, connection of primary particles proceeds suitably at the radially outer portion of the secondary particle, but extremely at the center area of the secondary particle, thereby forming secondary particle having too large connection areas among secondary particles.

Accordingly, in case of secondary particle which is composed of fine primary particles, which are arranged in radial direction outwardly, it is difficult to control the contact areas among primary particles.

The contact areas among the primary particles may vary in a secondary particle.

In case of LiCoO_2 type positive electrode material of Matsumoto et al., each of the secondary particles includes such primary particles having relatively too large contact areas partly or locally. If the volume change of secondary particles due to charging/discharging occurs, it causes cracking along the boundary of such primary particles having relatively too large contact areas. Accordingly, life span of battery becomes short, and it is difficult to maintain the battery capacity.

The Matsumoto et al. publication discloses that at least a part of primary particles which are arranged in radial direction are connected by sintering, the electron conductivity can be improved.

The fine primary particles have many fine gaps in which the electrolyte can infiltrate.

In such structure, electron conductivity may decrease due to gaps. Accordingly, in order to improve electron conductivity, at least a part of primary particles are connected by sintering.

Since the primary particles of Matsumoto et al. are arranged in radial directions in secondary particle, gaps among primary particles are formed resulting in deterioration of electron conductivity. To improve electron conductivity, at least a part of primary particles are connected by sintering. The Matsumoto et al. publication discloses that electron conductivity can be improved by sintering, but fails to teach increasing the contact areas among primary particles.

The Matsumoto et al. publication also discloses in [0013] that pack density can be raised but fails to teach increasing the contact areas among primary particles. Accordingly, one skilled in the art could not have made the present invention even if Matsumoto is combined with Shiozaki (WO 03/044881).

The Shiozaki et al. publication discloses a positive electrode active material for a lithium secondary cell having a high energy density and excellent in charging/discharging cycle performance, and a lithium secondary cell having a high energy density and excellent in charging/discharging cycle performance are disclosed. A positive electrode active material for a lithium secondary cell is characterized in that the composition of the active material is $\text{Li}_x\text{Mn}_a\text{Ni}_b\text{Co}_c\text{O}_2$ (where a, b, and c are values of a point (a, b, c) on a side of or inside a quadrilateral ABCD having vertexes A(0.5, 0.5, 0), B(0.55, 0.45, 0), C(0.55, 0.15, 0.30), and D(0.15, 0.15, 0.7) on a ternary state diagram showing the relationship among a, b, and c, and satisfy the expressions $0.95 < x/(a+b+c) < 1.35$) and in that the active material contains a composite oxide having an $\alpha\text{-NaFeO}_2$ structure. The lithium secondary cell comprises this active material.

By determining only the voidage of secondary particle, it is not possible to avoid primary particles which have small contact areas among primary particles. In manufacturing positive electrode material, such secondary particles which include primary particles having relatively small contact areas are inevitably produced.

To improve electron conductivity, it is necessary to avoid such primary particles having small contact areas.

According to the present invention, “length in which the primary particles are linked on the section of the secondary particle is equivalent to 10 to 70% of the length of the whole periphery on the section of the primary particles” and “the voidage of the secondary particle is 2.5 to 35 %.” Further, according to the present invention, the percentage content of Ni (y) is less than 50%, so that the relative lattice volume change rate decreases.

As described above, the present invention includes three features. By satisfying those three features, a lithium secondary battery using the positive electrode material in which the discharge rate characteristic and the battery capacity in the environment of the low temperature of -30°C are hardly deteriorated and which are excellent in a cycle characteristic can be achieved. One skilled in the art could not have thought the present invention having those three features based on Matsumoto et al. and Shiozaki et al. The object of the present invention is different from that of Shiozaki et al. The Shiozaki et al. publication does not teach the contact area among primary particles in each secondary particle of the present invention.

For the foregoing reasons, the presently claimed invention is patentable over the proposed combination of Matsumoto et al. and Shiozaki et al.

In view of the foregoing amendments and remarks, favorable reconsideration and allowance of all of the claims now in the application are requested.

To the extent necessary, applicants petition for an extension of time under 37 CFR 1.136. Please charge any shortage in the fees due in connection with the filing of this paper, including extension of time fees, to the deposit account of Antonelli,

Terry, Stout & Kraus, LLP, Deposit Account No. 01-2135 (Case: 1021.43559X00),
and please credit any excess fees to such deposit account.

Respectfully submitted,

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